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Hybrid seawater desalination-carbon capture using modified seawater battery system

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HIGHLIGHTS

- A Hybrid-type Seawater Desalination-Carbon Capture system is demonstrated.
- The hybrid system uses NASICON membranes to utilize Na^+ ions from seawater.
- Charging of the system can remove Na^+ and Cl^- ions from seawater.
- Discharging of the system can capture CO_2 via formation of carbonate precipitates.

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ABSTRACT

The water and carbon cycles are central to the Earth's ecosystem, enabling the sustainable development of human societies. To mitigate the global issues of water shortages and climate change, we report a new electrochemical system that fulfills two functions—seawater desalination and carbon dioxide air-capture—during the charge and discharge processes. The seawater desalination-carbon capture system utilizes a seawater battery platform, consisting of three major compartments (desalination, sodium-collection, and carbon-capture), which are separated by sodium superionic conducting ceramic membranes. It is found that the concentrations of sodium ions and chloride ions in fresh seawater (total dissolved solids ≈ 34,000 ppm) are significantly decreased by the charging of the seawater desalination-carbon capture system, resulting in brackish water (total dissolved solids ≈ 7000 ppm). The discharge process induces the air-capture of ambient carbon dioxide gases through carbonation reactions, which is demonstrated by the carbon dioxide gas removal in this compartment. The hybrid system suggests a new electrochemical approach for both desalination and carbon capture.

1. Introduction

As the planet's most abundant resource, seawater accounts for 70% of the Earth's surface, contributing to various natural cycles in the Earth ecosystem. One representative cycle is the water cycle, in which solar energy evaporates seawater to separate salts and water vapor, producing fresh water [1]. Another significant cycle is the carbon cycle, which maintains the atmospheric carbon dioxide (CO_2) level by capturing the gas in seawater and forming carbonated materials such as $CaCO_3$ and $CaMg(CO_3)_2$ [2]. However, the sharp increase in the world population and the use of fossil fuels have caused water scarcity [3–5] and the elevation of the CO_2 concentration in the atmosphere. According to the Worldwatch Institute, an estimated 1.8 billion people will reside in complete water scarcity regions in 2025, while almost 50% of the world's population will suffer from water stresses [6]. Additionally, the Earth is naturally able to process 14 billion tons of CO_2 per year by diffusing CO_2 into water or by photosynthesis by green plants [7]. However, according to the report [7], the CO_2 emissions in 2050 are predicted to be 62 billion tons per year, which is more than four times the level that the Earth can handle per year.

Such significant environmental issues have led to development of various technologies. For such as evaporation at elevated temperatures or filtration through reverse osmosis membranes for anthropogenic desalination [8–21] and carbon capture and storage (CCS) technologies

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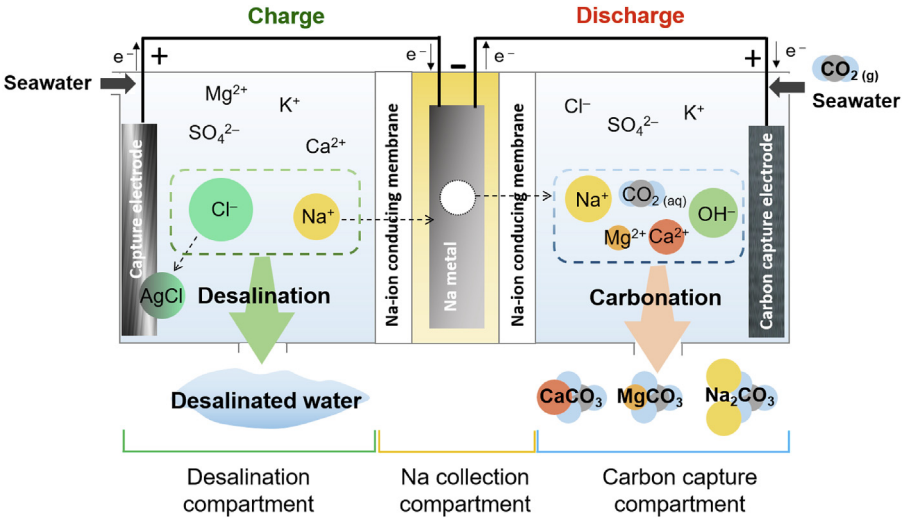

Fig. 1. Schematic representation of the hybrid desalination-carbon capture system (SD-CC system). By charging the system, Na^+ and Cl^- ions from seawater are separated into the collection and capture compartments, respectively. Upon discharging, the SD-CC system captures CO_2 gases from the ambient air through carbonization in seawater, while producing electricity to external loads.

for the reduction of CO_2 emissions [22–32]. Nevertheless, such approaches still require improvement in terms of energy efficiency, operation cost, and environmental compatibility. For example, the desalination technologies, reverse osmosis, requires a large electrical energy input and thermal energy based multistage flash distillation requires energy intensive heating to temperatures above 90 °C [16]. And for reduction of carbon capture, typical CCS technologies are based on stationary, massive infrastructures, which require long installation time and intensive management, eliminating CO_2 gases only within limited areas [25,29].

In this study, we introduce a novel electrochemical device that can complete both seawater desalination and CO_2 capture at room temperature by the charging and discharging of the device, individually. The idea for this device, termed a seawater desalination–carbon capture (SD-CC) system, arose from the discovery of by-products generated during operation of rechargeable seawater batteries [33–40]. A seawater battery utilizes Earth-abundant seawater as the active material in an open-structured cathode. This battery uses a Na super ion conductor (NASICON) solid electrolyte to physically separate the anode and cathode compartments of the cell, allowing only Na-ion transport between the seawater cathode and the anode. The charging process of the seawater battery results in the decrease of NaCl (*aq*) concentration in the seawater, whereas the discharging generates by-products as metal carbonates. By capitalizing on these reactions, we extended a seawater battery platform to construct a hybrid SD-CC system.

The hybrid SD-CC system was designed to consist of three major compartments dedicated to desalination, Na-collection, and carbon capture. In this work, the Cl^- ion-capture electrode was used in the desalination compartment to efficiently extract Cl^- ions from the seawater. By charging the SD-CC cell, the concentration of Na^+ and Cl^- ions in fresh seawater (total dissolved solids, TDS ~ 34,000 ppm) was significantly decreased to TDS ≈ 7000 ppm (brackish water). During the discharging of the SD-CC cell, the Na metal in the Na-collection compartment is oxidized into Na^+ ions, which are released into the seawater, while forming hydroxide (OH^-) ions simultaneously. Subsequently, the hydroxide ions and Na^+, Ca^{2+}, and Mg^{2+} cations in seawater react with CO_2 (*aq*) introduced from the ambient atmosphere to form metal carbonates such as Na_2CO_3, $CaCO_3$, and $MgCO_3$. We investigated such bi-functionality and the electrochemical reactions behind the cell operation using various analytical techniques. In addition, the consecutive charge and discharge processes were performed to evaluate the viability of the SD-CC system, demonstrating both functions of seawater desalination and CO_2 -capture at room temperature for the first time. With this new design, our system would provide a great potential for both desalination and CO_2 capture technologies and contribute to restoring natural cycles of the Earth.

2. Materials and methods

2.1. Preparation of the SD-CC cell components

The SD-CC system is based on a seawater battery configuration. NASICON ceramic membranes ($Na_{1+x}Zr_2Si_xP_{3-x}O_{12}$, x = 2), with 16 mm in diameter, were fabricated according to the previous report [33]. X-ray diffraction (XRD) was carried out to identify the NAISICON phase (Fig. S1). The ceramic membrane was mechanically polished down to a thickness of 0.8 mm before the cell assembly. An organic electrolyte, 1 M $NaCF_3SO_3$, dissolved in tetraethylene glycol dimethyl ether (TEGDME) solvent, was used in the Na-collecting compartment. A nickel mesh (Alfa Aesar) was used as an anode current collector. A polyethylene separator with a thickness of 25 μm (Celgard) was inserted as a buffer layer between the NASICON and the organic electrolyte. A 25 μm-thick silver foil (Alfa Aesar, 10 mm in diameter) was used as a Cl^--capture electrode for desalination. The Ag foil was polished by an abrasive cloth (Deer KA161, 100 grit) to roughen the surface before assembling the cells. A carbon paper with a thickness of 370 μm and an electrical resistivity of less than 20 Ω-mm was used as a cathode current collector for CO_2 capture. Natural seawater from Ilsan Beach, Ulsan, Republic of Korea, was used after filtration, and its quantitative information can be found elsewhere.[24]

2.2. Fabrication of the SD-CC cell

The SD-CC system consists of three distinct compartments: a desalination compartment, the Na-collection compartment, and a carbon capture compartment, as schematically illustrated in Fig. 1. NASICON membranes were mounted on both sides of the Na-collection compartment, separating the other two compartments. The Na-collection compartment was assembled in an argon-filled glovebox using a Ni mesh as the current collector and polyethylene separators (Celgard) soaked with 1 M $NaCF_3SO_3$ (TEGDME) electrolyte were inserted as a buffer layer between the Ni mesh and NASICON before the sealing the compartment. The assembled Na-collection compartment was inserted between the desalination and CO_2 capture compartments. The desalination compartment used an Ag foil to extract Cl^- ions present in seawater through the formation of AgCl particles on the surface of the foil. After the desalination process (system charging), the desalinated seawater was removed into a storage tank. The CO_2 capture compartment employed a porous carbon paper electrode to generate OH^- ions while the system discharged. The carbon capture compartment was

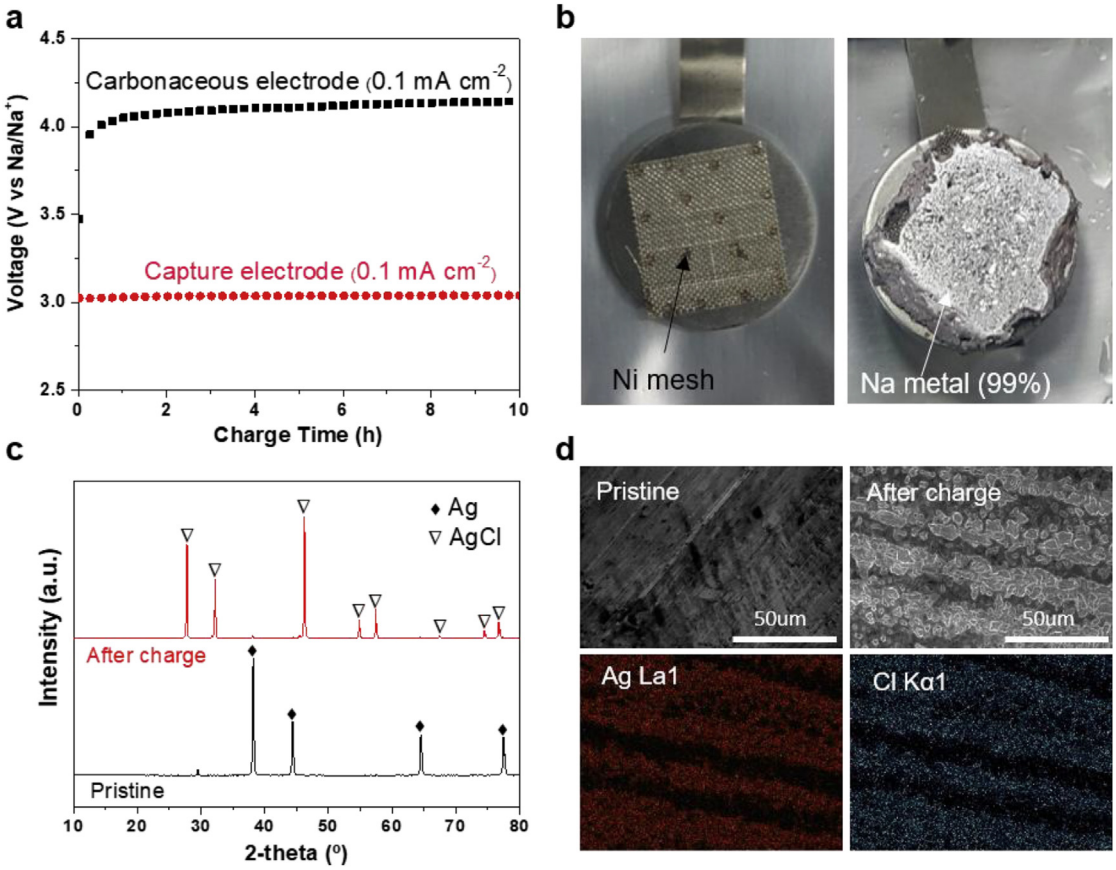

Fig. 2. (a) Voltage profiles of the SD-CC system with different electrodes during charge at a current condition of 0.1 mA cm^{-2}; (b) digital images of Na metal deposited on the collection compartment before and after the charge; (c) XRD patterns of the capture electrode (Ag foil) before and after the charge; and (d) SEM (top) and EDS mapping images (bottom) of the Ag foil before and after the charge.

placed in a completely sealed glass container, where CO_2 gas was maintained at a level of 500 ppm, and a CO_2 gas sensor was mounted in the sealed container to monitor the CO_2 concentration. The resulting seawater containing metal carbonate precipitates were subjected to filtration yielding a powder comprised of carbonated materials.

2.3. Characterizations

Electrochemical tests were performed with an automatic galvanostatic unit (WonATech. Co.). The purity of the extracted metal was determined by inductively coupled plasma–mass spectrometry (ICP-MS, 700-ES, Varian Inc). The surface morphology was observed by field-emission scanning electron microscopy (SEM, S-4200, Hitachi). The elemental distribution was examined by energy dispersive X-ray spectroscopy (EDS, Bruker Xflash 6130). The ion concentration was estimated by ion chromatography (IC, Dionex ICS 3000 and Dionex ICS 1600, Thermo Fisher Scientific). The X-ray diffraction (XRD) pattern was obtained with an X-ray diffractometer (D/Max 2000, Rigaku) in the 2θ range of 10–80° with a step interval of 0.5°. The CO_2 concentration and pH were monitored by using an ultralow power carbon dioxide sensor (COZIR™, GSS) and a pH meter (ORION STAR A216, Thermo Fisher Scientific), respectively.

3. Results and discussion

The SD-CC system operates in a similar manner to the charge and discharge processes of seawater batteries. When the SD-CC system is charged by electric energy, Na^+ ions are moved from seawater into the Na-collection compartment through the NASICON, followed by Na deposition occurred at the anode electrode (Equation (1)). While at the cathode-side, when using the carbon electrode, oxygen and/or chlorine evolution reaction occurs (Equations (2) and (3), refer to Fig. S2). But, for better desalination process, it is desired chloride ions by promoting the chlorine evolution reaction rather than oxygen evolution reaction in seawater. Therefore, it is compulsory to use a specific catalyst to promote the chlorine evolution reaction. However, it is quite difficult at present condition to promote a chlorine evolution reaction because the system still needs further an engineering management and environment modification at cathode side of the system. Therefore, presently to prove the concept of desalination in SD-CC system, temporarily Ag foil is employed as the chloride (Cl^-)-ion-capturing electrode. When using Ag foil, the Cl^--ion-capturing is occurs at the cathode by the formation of AgCl on the Ag foil (Equation (4)), which is different from the cathodic reaction of seawater batteries (Equation (2)). Consequently, Na^+ and Cl^- ions are extracted from seawater into each compartment during the charging of SD-CC system, generating desalinated water ('desalination process'). The half-cells for the corresponding electrode potentials and the overall reaction of the SD-CC system are given below.

$$Na^+ + e^- \leftrightarrow Na;\ E^o = -2.71\ V\ vs.\ NHE \quad (1)$$

$$O_2 + 2H_2O + 4e^- \leftrightarrow 4OH^-\ (aq);\ E^o = 0.75\ V\ vs.\ NHE\ (pH = 8) \quad (2)$$

$$2Cl^- \rightarrow Cl_2 + 2e^-;\ E^o = +1.36\ V\ vs.\ NHE \quad (3)$$

$$Ag\ (s) + Cl^-\ (aq) \leftrightarrow AgCl\ (s) + e^-;\ E^o = 0.222\ V\ vs.\ NHE \quad (4)$$

$$4Na + O_2 + 2H_2O \leftrightarrow 4NaOH\ (aq);\ E^o = 3.46\ V\ vs.\ Na/Na^+\ (pH = 8) \quad (5)$$

$$Ag(s) + Na^+ + Cl^-\ (aq) \leftrightarrow AgCl\ (s) + Na;\ E^o = 2.932\ V\ vs.\ Na/Na^+ \quad (6)$$

To demonstrate the desalination process, we assembled two different cells for comparison: one employed a carbon paper electrode, corresponding to an existing seawater battery system (refer to Fig. S2), and the other employed an Ag foil electrode to capture the chloride ions (SD-CC system). We then charged the two cells at a constant current of 0.1 mA cm^{-2}. The cell using the carbon electrode (black curve) showed a voltage plateau of 4.15 V. Compared to the theoretical value (3.46 V vs. Na/Na^+), the high voltage plateau for the carbon electrode cell resulted from a large overpotential induced from sluggish kinetics of the oxygen evolution reaction in seawater (Equation (5)) [41,42]. On the other hand, the Ag electrode cell (red curve) showed a voltage plateau (~3.04 V vs. Na/Na^+) close to the theoretical voltage (Equation (6)). It should be noted that the Na-collection compartments of both cells commonly accompany the Na deposition reaction (Equation (1)) during charging. After disassembling the charged cells, we observed Na metal deposited on the Ni mesh current collector in the Na-collection compartment, as shown in Fig. 2b. The ICP-MS analysis verified that the collected deposit was Na of ~99% purity with a trace of S impurity (~1%), which was probably due to the liquid electrolyte of $NaCF_3SO_3$ (in TEGDME). We also performed XRD of the Ag electrode after charging. As displayed in Fig. 2c, the XRD pattern had a major phase of cubic AgCl (space group: $Fm\bar{3}m$ (225), JCPDS#31–1238) and a minor phase of cubic Ag (space group: $Fm\bar{3}m$ (225), JCPDS #04–0783), which agrees with the SEM-EDS results, showing that the AgCl particles uniformly covered the Ag surface (Fig. 2d). These results indicate that the Cl^- and Na^+ ions from seawater were extracted into the Ag electrode and Na-collection compartments during the charging process, respectively.

To estimate the concentrations of remaining Na^+ and Cl^- ions in the desalinated seawater after charging, we conducted IC on the processed seawater as a function of charging time. Fig. 3 displays the variations of Na^+ (a) and Cl^- (b) ion concentrations with charging time at different current densities from 0.1 to 0.5 mA cm^{-2}. It is noteworthy that as the charging time increased, the concentrations of the Na^+ and Cl^- ions decreased. Although there are small deviations on the graphs, the higher applied current density induced rapid decreases in the ion concentration with increasing charging time. For example, the ion concentrations of Na^+ and Cl^- in the seawater after 8 h charging at 0.1 mA cm^{-2} were reduced by as much as 3.3% and 8.3%, respectively. On the other hand, charging at 1.0 mA cm^{-2} for the same time resulted in decreases of 19.9% and 20.4%, respectively.

We constructed bar charts to compare the concentrations of ion species present in seawater, including Na^+ and Cl^- ions, before and after the charging process. As shown in Fig. S3, when 25% and 50% of Cl^- ions were removed by the charging, the Na^+ ion concentration decreased almost proportionally to the degree of Cl^- ion removal. On the other hand, the concentration of the other ion species appeared to be almost unchanged, regardless of the charging process. This indicates that the charging process of the SD-CC system selectively removes Na^+ and Cl^- ions in seawater. We charged the SD-CC system loaded with 4 mL of fresh seawater until the Na^+ ions were almost removed. Table 1 shows the ion concentration in the seawater before and after charging the SD-CC system. The fresh seawater had a total dissolved solids (TDS) concentration of ~34,000 ppm, which was comparable to that of standard seawater (~35,000 ppm) [44,45]. After charging at 0.1 mA cm^{-2} for 120 h, the TDS of the seawater was reduced to 7000 ppm, which means that 80% of TDS was removed from the pristine seawater. The reduced ionic species were mostly Na^+ and Cl^- ions, which confirms that the charging process of the SD-CC system selectively eliminates Na^+ and Cl^- ions in seawater. According to the previous report by Moulin et al., water with a TDS concentration ranging from 3000 ppm to 10,000 ppm is classified as brackish water [46]. Therefore, the SD-CC system can produce desalinated water (~7000 ppm) comparable to brackish water.

As the second function, the air-capture of CO_2 results from a carbonation process at work during the discharging of the SD-CC system,

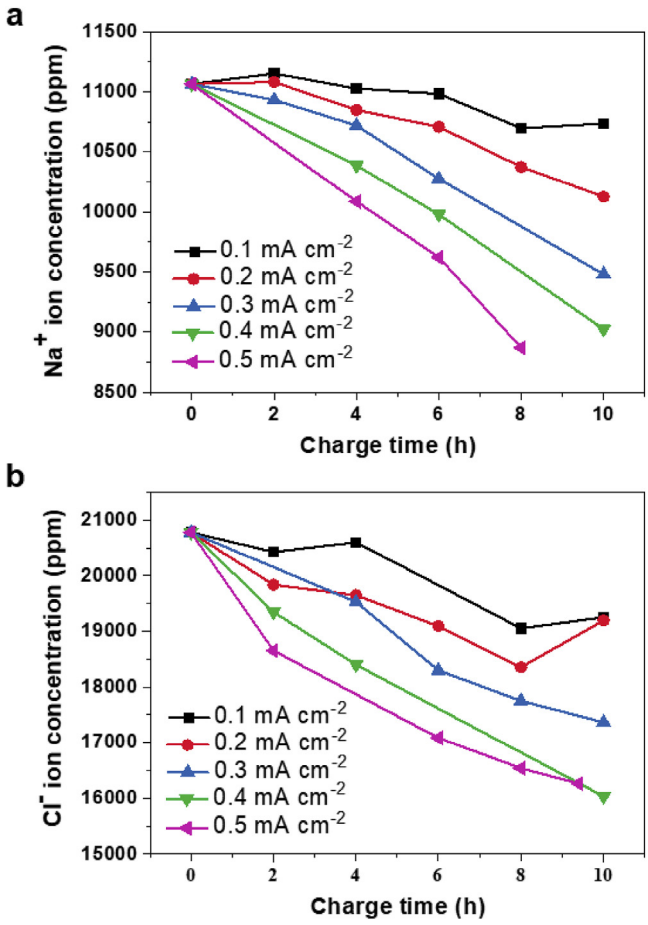

Fig. 3. Changes of the concentration of (a) Na^+ and (b) Cl^- ions with various charge current densities of 0.1–0.5 mA cm^{-2} for 10 h.

Table 1
Ion concentrations and TDS before and after the charge of SD-CC system with the Ag capture electrode.

	Ions (ppm)						
	Na	Mg	K	Ca	Cl	SO_4	TDS
Pristine Seawater	11,000	1300	500	400	18,000	2400	34,000
After charging	300	1200	300	400	2400	2400	7000

as depicted on the right side of Fig. 1. At the Na-collection compartment, the Na metal is oxidized into Na^+ ions (Equation (1)), which are transported into the seawater through the NASICON membrane, while the oxygen reduction reaction (ORR) occurs at the carbon electrode (Equation (2)), powering external loads. The dissolved oxygen for the oxygen reduction reaction (ORR) is supplied from the ambient air. Owing to the open-structured cathode configuration, the SD-CC system can utilize the oxygen continuously, in order to form NaOH (aq) during discharging. As the discharge process proceeds, the seawater pH near the carbon electrode is increased by the increased amount of OH^- ions. A portion of the NaOH (*aq*) would react with Ca^{2+} and Mg^{2+} present in seawater, forming metal hydroxides (*aq*), as described in Equations (7) and (8) [43].

$$2NaOH\ (aq) + MgCl_2\ (aq) \rightarrow 2NaCl\ (aq) + Mg(OH)_2\ (aq) \quad (7)$$

$$2NaOH\ (aq) + CaCl_2\ (aq) \rightarrow 2NaCl\ (aq) + Ca(OH)_2\ (aq) \quad (8)$$

As long as the discharge process continues, OH^- ion concentration increases, and the metal hydroxides (*aq*) could act as absorbents to capture ambient CO_2 gases [44]. The overall reactions of CO_2 (g) absorption can be expressed as in Equations (9) and (10). Therefore, the

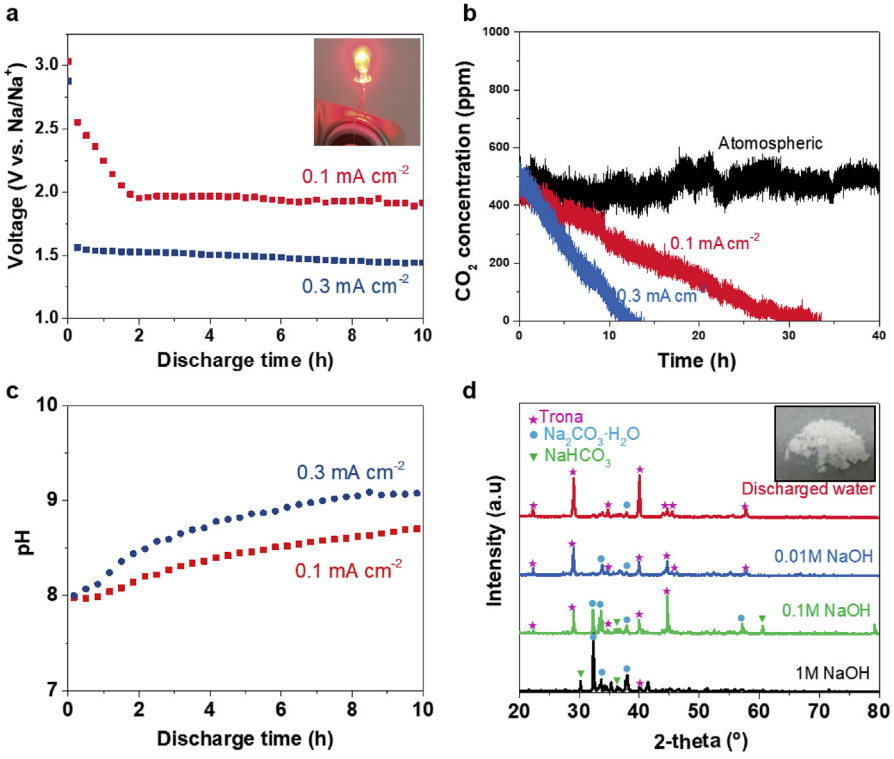

Fig. 4. (a) Voltage profiles of the SD-CC system during the discharge process at current conditions of 0.1 mA cm^{-2} and 0.3 mA cm^{-2}. The inset shows an LED powered by the discharge process. (b) Concentration changes of the CO_2 during discharge process at 0.1 mA cm^{-2} (red) and 0.3 mA cm^{-2} (blue). The initial CO_2 concentration level in the SD-CC system was set as 500 ppm, comparable to the atmospheric CO_2 level (black). (c) The pH profiles of seawater during discharge process at current conditions of 0.1 mA cm^{-2} and 0.3 mA cm^{-2}; (d) XRD patterns of the dried powders after discharge-induced carbonation process at 0.1 mA cm^{-2} using a DI water and NaOH (*aq*) with various concentrations of 0.01 M–1 M. The inset shows the acquired powder from the DI water by discharging. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

discharge process enables the system to not only provide electricity to external loads, but also to capture ambient CO_2 gases through the formation of metal carbonates in seawater. Unlike the previous methods of CO_2 capture in NaOH solution [47–49], which need a continuous supply of NaOH solution, our system produces NaOH (*aq*) during discharging, eliminating the price of the raw material of NaOH, which is produced by energy-requiring reactions such as chlor-alkali reaction [50,51].

$$2NaOH\ (aq)\ +\ CO_2\ (g) \rightarrow Na_2CO_3\ (aq)\ +\ H_2O\ (l) \quad (9)$$

$$M(OH)_2\ (aq)\ +\ 2CO_2\ (g) \rightarrow MCO_3\ (aq)\ +\ H_2O\ (l);\ M = Mg \text{ or } Ca \quad (10)$$

The carbonation process during the SD-CC system discharging was investigated under current conditions of 0.1 mA cm^{-2} and 0.3 mA cm^{-2} for 10 h. Fig. 4a shows the voltage profiles for the discharge process. The discharging produced electricity for the external load (lighting of a red LED bulb) as displayed in the inset of Fig. 4; the high current (at 0.3 mA cm^{-2}) accompanied lower discharge potential than that for the 0.1 mA cm^{-2}, due to the higher overpotential of the ORR [34,37,38,40]. We examined the CO_2 air-capture of seawater during the discharge process. To examine the CO_2 air-capture, the carbon capture compartment containing seawater was completely sealed in a glass container, where the initial level of CO_2 was maintained at 500 ppm (as indicated by the black curve in Fig. 4b), and a CO_2 gas sensor was present to monitor the CO_2 concentration. As shown in Fig. 4b, the discharge process resulted in the monotonic decrease of the CO_2 concentration with longer discharge times; the higher applied current (0.3 mA cm^{-2}) reduced the CO_2 level more rapidly than the low current condition (at 0.1 mA cm^{-2}) did. This result verifies that the discharging of the SD-CC system induces air-capture of CO_2 gases. As we mentioned above, the discharging produces OH^- ions in seawater due to the ORR on the carbon electrode (Equation (2)). To verify this, we measured the pH change of seawater during the carbonation process *in-situ* while discharging at 0.1 mA cm^{-2} and 0.3 mA cm^{-2}. As shown in Fig. 4c, a faster increase in the pH was found at the higher current of 0.3 mA cm^{-2} than at the lower current for the same discharging time, which indicates a high yield of OH^- ions in the carbon-capture compartment, enabling a relatively high efficiency of CO_2 capture per unit time, which agrees with the results of Fig. 4b.

To confirm the CO_2 air-capture via the carbonation during discharging, we conducted control experiments, where 20 mL of DI water was used as the absorber electrolyte in the carbon capture compartment. As shown in Fig. S4, we performed the discharge process at current densities of 0.1 mA cm^{-2} and 0.3 mA cm^{-2} for 300 h. Afterwards, the concentrated DI water for 0.3 mA cm^{-2} was dried at 35 °C in ambient air for a few days, in order to accelerate the formation of precipitates and analyse the phase (see the inset of Fig. 4d). After discharging for 100 h, the DI water was dried at 35 °C in ambient air for a few days, producing white precipitates (see the inset of Fig. 4d). For comparison, air-capture tests of CO_2 were carried out not by discharging, but by using aqueous NaOH solutions with various concentrations between 0.01 M and 1 M, and the resulting precipitates were treated in the same manner as those obtained from discharging in the DI water. The phases of all the precipitates were identified by XRD. As can be seen in the XRD patterns of Fig. 4d, the white product was analysed as a major phase of trona ($Na_2CO_3{\cdot}NaHCO_3{\cdot}H_2O$), together with minor phases of $Na_2CO_3{\cdot}H_2O$ and/or $NaHCO_3$. For the 1 M NaOH solution, the precipitates consisted of mainly thermonatrite ($Na_2CO_3{\cdot}H_2O$), together with a small amount of $NaHCO_3$. This result reveals a typical carbonization process in the chemical reaction between NaOH (*aq*) and CO_2 (g), producing sodium (bi)carbonates (*aq*). As the concentration of the NaOH solution decreased from 1 M to 0.01 M, the amount of thermonatrite and $NaHCO_3$ phases was decreased, while that of the trona phase was increased. This result indicates that at lower alkalinity levels, a larger proportion of trona is found in the precipitates. It is noteworthy that the precipitates acquired after the discharge-induced carbonation process at a current of 0.1 mA cm^{-2} for 100 h had a major phase of trona, similar to the precipitates formed with the 0.01 M NaOH (*aq*). This confirms that the discharging of the SD-CC system resulted in CO_2 air-capture through a carbonation reaction of the catholyte, in which OH^- ions are continuously increased by the discharging process.

Next, we discharged the SD-CC system using seawater at a current of 0.1 mA cm^{-2} for 100 h, resulting in precipitation in the bottom of the

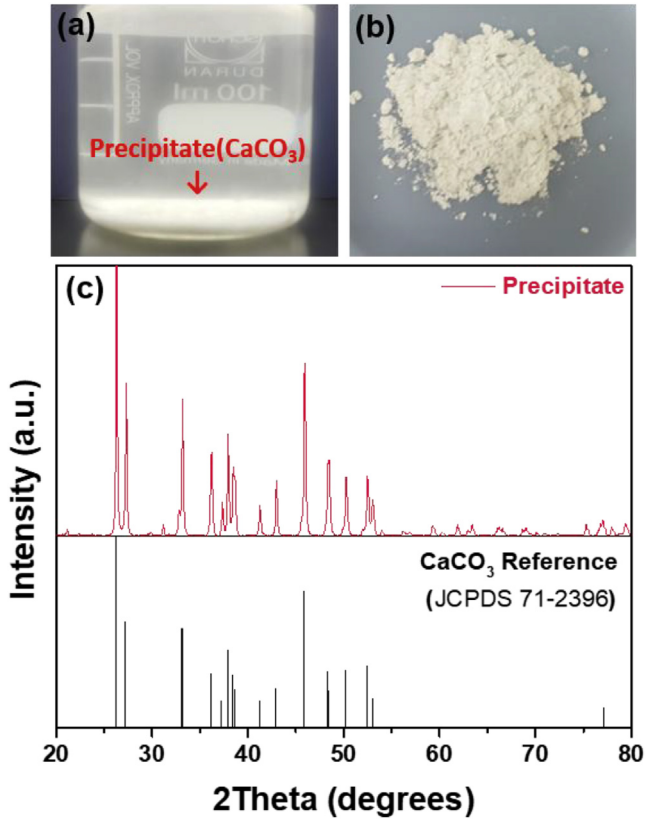

Fig. 5. Photographs of the precipitate in seawater (a) and dried one (b) after the discharge-induced carbonation process; (c) XRD pattern of the dried precipitate. The bottom panel shows the reference diffraction line for the $CaCO_3$ phase.

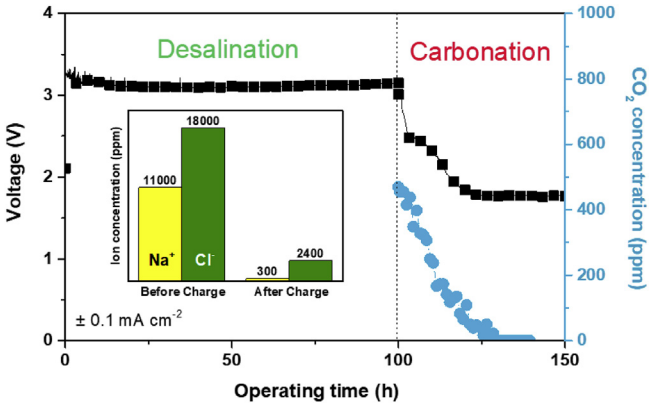

Fig. 6. Voltage profile of the SD-CC system during successive charge and discharge processes. The inset bar chart shows the Na^+ and Cl^- ion concentrations before and after the charge process. The right blue graph indicates the variation of CO_2 gas concentration during the discharge-induced carbonation process. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

seawater (see Fig. 5a). The precipitates were prepared by centrifugation, filtering, and drying at 80 °C overnight (see Fig. 5b) and were analysed by XRD. Fig. 5c shows the XRD data of these white precipitates, which were phase-identified as $CaCO_3$. The formation of $CaCO_3$ in seawater appeared to be a result of the carbonation of Ca $(OH)_2$, as we mentioned previously (Equation (10)), which indicates a discharge-induced seawater carbonation process. Although the concentration of Mg^{2+} ion (~1.29 g/kg) is higher than that of Ca^{2+} ion (~0.41 g/kg) in a typical seawater, the formation of $CaCO_3$ is thermodynamically favored over that of $MgCO_3$ in seawater [52]. We performed a consecutive charge and discharge process of the SD-CC system using the Ag and carbon electrodes in the desalination compartment and carbon-capture compartment, respectively, with seawater at a current of 0.1 mA cm^{-2}, as shown in Fig. 6. The SD-CC system is a continuous processing system divided by three different compartments, as shown in Fig. 1. The system continues to charge, until the silver electrode is totally covered with AgCl in the desalination compartment. On the one hand, the discharge process continues to capture ambient CO_2 in the carbon capture compartment, until all sodium metal harvested in the Na collection compartment is oxidized (consumed). The subsequent discharging induced the nearly complete capture of CO_2 gases within 50 h via the carbonation of seawater in the SD-CC system. It should be noted that the relatively short processing time of discharging (< 50 h), compared to that of charging (100 h), was due to the limited amount of CO_2 gases in the sealed carbon capture compartment. These results show that the SD-CC system functions for both seawater desalination and CO_2 capture during the charge and discharge processes, respectively, and suggest a new hybrid-type bifunctional energy conversion/storage system to mitigate both energy and environmental issues.

4. Conclusions

We modified a rechargeable seawater battery system to add two functions, seawater desalination, and CO_2 capture, developing a new type of hybrid SD-CC system. We constructed three distinct compartments in the SD-CC system for desalination, Na-collection, and carbon-capture, which were separated by NASICON membranes. The charging of the SD-CC system led to the oxidation of Cl^- ions in seawater to form AgCl particles on the chloride-capture electrode (Ag foil). At the same time, Na^+ ions in seawater were transported into the Na-collection compartment through the NASICON membrane, followed by deposition of Na metal. As a result, Na^+ and Cl^- ions were removed from seawater, producing desalinated seawater (*desalination*). During the SD-CC system discharging, the Na metal in the Na-collection compartment was oxidized into Na^+ ions, which were moved into the carbon-capture compartment through the NASICON, while the ORR occurred at the carbon electrode, providing electric power to external loads. The generated OH^- ions induce the air-capture process of CO_2 gases from the ambient atmosphere by the formation of $CaCO_3$ (*carbonation*), due to the reaction with Ca^{2+} ions present in seawater. Consecutive charge and discharge processes of the SD-CC system at a current condition of 0.1 mA cm^{-2} resulted in the removal of 97.3% Na^+ and 86.7% Cl^- ions from fresh seawater and complete CO_2 air-capture within 50 h. However, it should be noted that the expensive Ag is used for temporarily to prove the concept of desalination using SD-CC system. This report provides the possible methodology to develop an SD-CC system for desalination and carbon-capture. We expect further development at the cathode side of SD-CC system for the continuous desalination process by the proper engineering modification and by employing efficient chlorine evolution catalyst-coated electrode. We hope that the proposed SD-CC system could provide the promising direction in the future to resolve energy and environmental issues, including electricity demand, water shortage, and climate change.

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Appendix A. Supplementary data

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